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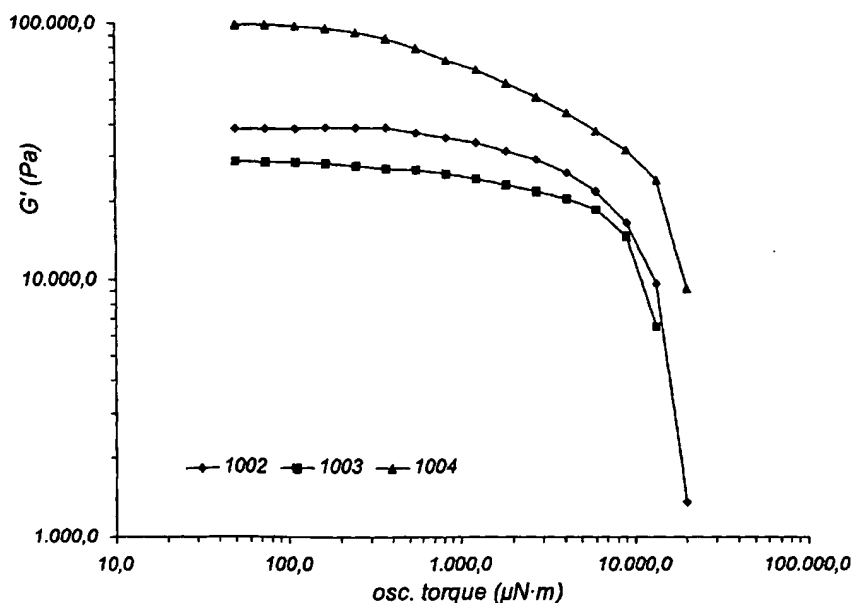
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(54) Title: BIODEGRADABLE CHEWING GUM COMPRISING AT LEAST ONE HIGH MOLECULAR WEIGHT  
BIODEGRADABLE POLYMER



(57) Abstract: The invention relates to a chewing gum comprising at least one biodegradable polymer, wherein the molecular weight of said polymer is at least 105000 g/mol (Mn). According to the invention, it has moreover been realized that this problem may be effectively dealt with by increasing the molecular weight of at least one of the biodegradable polymers in the chewing gum when compared to conventional chewing gum polymers and thereby increasing the robustness of the chewing gum with respect to softeners, emulsifiers and e.g. flavor.

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## BIODEGRADABLE CHEWING GUM COMPRISING AT LEAST ONE HIGH MOLECULAR WEIGHT BIODEGRADABLE POLYMER

### Field of the invention

5

The present invention relates to biodegradable chewing gum according to claim 1.

### Background of the invention

10 US patent 5,672,367 discloses a chewing gum made on the basis of a biodegradable polymer having polymeric properties, i.e. a polymer. A significant advantage, as explained in the patent, is that the resulting chewing gum may be degraded when disposed in the nature or e.g. on streets as a result of normal environmental influence. The disclosed chewing gum is claimed as having a structure and chewing  
15 characteristics comparable to those of chewing gum based on conventional, non-degradable polymers. It has moreover been realized, that the exchange of polymers provides a reduced tackness.

US patent 6,153,231 discloses a chewing gum comprising a biodegradable polymer  
20 and where the polymer comprises a poly(lactic acid-dimer fatty acid oxazoline) copolymers or a poly(lactic acid-diol-urethane) copolymer.

A problem of the above disclosed chewing gums is that the biodegradable polymers as such does not simply replace the polymer or polymers of conventional chewing  
25 gum. It has been realized that the simple polymer substitution results in an overall chewing gum having different characteristics than conventional chewing gum. This problem indicates that the substituted polymer reacts differently with the remaining chewing ingredients than the biodegradable polymers thereby rendering conventional chewing gum formulations more or less useless.

30

It is an object of the invention to provide a biodegradable chewing gum capable of incorporating at least certain important chewing gum ingredients.

### Summary of the invention

- 5 The invention relates to chewing gum comprising at least one biodegradable polymer, wherein the molecular weight of said at least one polymer is at least 105.000 g/mol (Mn).

10 According to the invention, it has been realized that chewing gums made on the basis of biodegradable polymers are somewhat vulnerable to different conventional chewing gum additives or components. Most critically, it has been realized that softeners, which are highly needed when obtaining the desired chewing gum texture, tend to dissolve the chewing gums even when applied in small amounts.

- 15 According to the invention, it has moreover been realized that this problem may be effectively dealt with by increasing of the molecular weight of at least one of the biodegradable polymers in the chewing gum when compared to conventional chewing gum polymers and thereby increasing the robustness of the chewing gum with respect to softeners, emulsifiers and e.g. flavor.

20 According to the invention, it has moreover been realized that an increasing of the molecular weight of at least one of the biodegradable polymers and thereby an increasing of the rheological stiffness ( $G'$ ) may in fact be more than compensated by addition of softeners.

25 In other words, according to the invention an improved texture of a biodegradable polymer containing chewing gum may in fact surprisingly be obtained by an initial worsening of the rheological properties of the biodegradable polymer and finally be more than compensated by the addition of suitable softeners.

30 Due to the hydrophilic nature of biodegradable polymers, the polymers tends to swallow water, e.g. from mouth induced saliva. Thereby, the intermolecular forces

between the neighboring molecular chains will decrease and the chewing gum structure will weaken.

According to the invention, a higher resistance to the decreasing of intermolecular forces has been obtained partly due to the fact that the resulting intermolecular forces are increased between the polymer chain and moreover due to the fact that the increasing of the size of the molecular chains results in increased entanglement between the polymer chains of neighboring polymers.

10 According to the invention, it has moreover been realized that an improved long-term release of chewing gum ingredients may be obtained, with increased molecular weight of the applied biodegradable polymer.

In an embodiment of the invention the molecular weight of said at least one  
15 biodegradable polymer is at least 150000 g/mol ( $M_n$ ).

In an embodiment of the invention the molecular weight of said at least one biodegradable polymer is within the range of 105000 g/mol ( $M_n$ ) to 1000000 g/mol ( $M_n$ ).

20

According to the invention, it has been concluded that a sufficient amount of softeners may be added to a biodegradable polymer having a molecular weight of  $M_n = 113,900$  g/mol.

25 In an embodiment of the invention the molecular weight of said at least one biodegradable polymer is within the range of 105000 g/mol ( $M_n$ ) to 500000 g/mol ( $M_n$ ).

In an embodiment of the invention the molecular weight of said at least one  
30 biodegradable polymer is within the range of 105000 g/mol ( $M_n$ ) to 350000 g/mol ( $M_n$ ).

In an embodiment of the invention the molecular weight of said at least one biodegradable polymer is within the range of 105000 g/mol (Mn) to 250000 g/mol (Mn).

- 5 In an embodiment of the invention the molecular weight of said at least one biodegradable polymer is less than 2000000 g/mol (Mn).

In an embodiment of the invention the polydispersity of said at least one biodegradable polymer is within the range of 1 to 5.

10

In an embodiment of the invention the polydispersity of said at least one biodegradable polymer is within the range of 1 to 2,5 (2½).

- 15 In an embodiment of the invention the at least one biodegradable polymer comprises at least 25% of the chewing gum polymers, preferably at least 50%.

In an embodiment of the invention all the biodegradable polymers comprised in the chewing gum comprise at least 25%, preferably at least 50% of the chewing gum polymers.

20

In an embodiment of the invention all the biodegradable polymers comprised in the chewing gum comprise at least 80%, preferably at least 90% of the chewing gum polymers.

- 25 In an embodiment of the invention the chewing gum is substantially free of non-biodegradable polymers

In an embodiment of the invention said chewing gum ingredients comprise flavoring agents.

30

In an embodiment of the invention said flavoring agents comprise natural and synthetic flavourings in the form of natural vegetable components, essential oils,

essences, extracts, powders, including acids and other substances capable of affecting the taste profile

In an embodiment of the invention said chewing gum comprise flavor in an amount  
5 of 0.01 to about 30 wt%, said percentage being based on the total weight of the chewing gum

In an embodiment of the invention said chewing gum comprising flavor in an amount of 0.2 to about 4 wt%, said percentage being based on the total weight of the  
10 chewing gum

In an embodiment of the invention said flavor comprises water soluble ingredients.

In an embodiment of the invention said water soluble flavor comprises acids.  
15

According to the invention, a surprising initial release of acids has been obtained.

In an embodiment of the invention said flavor comprises water insoluble ingredients.

20 In an embodiment of the invention, said chewing gum ingredients comprise sweeteners.

In an embodiment of the invention said sweetener comprises bulk sweeteners

25 In an embodiment of the invention the chewing gum comprises bulk sweeteners in an amount of about 5 to about 95% by weight of the chewing gum, more typically about 20 to about 80% by weight of the chewing gum.

In an embodiment of the invention the sweetener comprises high intensity sweeteners  
30

In an embodiment of the invention the high intensity sweeteners comprise sucralose, aspartame, salts of acesulfame, alitame, saccharin and its salts, cyclamic acid and its

salts, glycyrrhizin, dihydrochalcones, thaumatin, monellin, sterioside, alone or in combination

In an embodiment of the invention wherein the chewing gum comprises high  
5 intensity sweeteners in an amount of about 0 to about 1% by weight of the chewing gum, more typically about 0.05 to about 0.5% by weight of the chewing gum.

In an embodiment of the invention, the chewing gum comprises at least one softener.

10 In an embodiment of the invention, the at least one softener comprises tallow, hydrogenated tallow, hydrogenated and partially hydrogenated vegetable oils, cocoa butter, glycerol monostearate, glycerol triacetate, lecithin, mono-, di- and triglycerides, acetylated monoglycerides, fatty acids, such as stearic, palmitic, oleic and linoleic acids mixtures thereof.

15

In an embodiment of the invention the chewing gum comprises softeners in an amount of about 0 to about 18% by weight of the chewing gum, more typically about 0 to about 12% by weight of the chewing gum.

20 In an embodiment of the invention, the chewing gum ingredients comprise active ingredients.

In an embodiment of the invention, said active ingredients are selected from the group of: Acetaminophen, Acetylsalicylsyre Buprenorphine Bromhexin Celcoxib  
25 Codeine, Diphenhydramin, Diclofenac, Etoricoxib, Ibuprofen, Indometacin, Ketoprofen, Lumiracoxib, Morphine, Naproxen, Oxycodon, Parecoxib, Piroxicam, Pseudoefedrin, Rofecoxib, Tenoxicam, Tramadol, Valdecoxib, Calciumcarbonat, Magaldrate, Disulfiram, Bupropion, Nicotine, Azithromycin, Clarithromycin, Clotrimazole, Erythromycin, Tetracycline, Granisetron, Ondansetron, Prometazin,  
30 Tropisetron, Brompheniramine, Ceterizin, leco-Ceterizin, Chlorcyclizine, Chlorpheniramin, Chlorpheniramin, Difenhydramine, Doxylamine, Fenofenadin, Guaifenesin, Loratidin, des-Loratidin, Phenyltoloxamine, Promethazin, Pyridamine,

- Terfenadin, Troxerutin, Methyldopa, Methylphenidate, Benzalcon. Chloride, Benzeth. Chloride, Cetylpyrid. Chloride, Chlorhexidine, Ecabet-sodium, Haloperidol, Allopurinol, Colchicine, Theophylline, Propanolol, Prednisolone, Prednisone, Fluoride, Urea, Miconazole, Actot, Glibenclamide, Glipizide,
- 5 Metformin, Miglitol, Repaglinide, Rosiglitazone, Apomorphin, Cialis, Sildenafil, Vardenafil, Diphenoxylate, Simethicone, Cimetidine, Famotidine, Ranitidine, Ratinidine, cetirizin, Loratadine, Aspirin, Benzocaine, Dextrometorphan, Ephedrine, Phenylpropanolamine, Pseudoephedrine, Cisapride, Domperidone, Metoclopramide, Acyclovir, Dioctylsulfosucc., Phenolphthalein, Almotriptan, Eletriptan, Ergotamine,
- 10 Migea, Naratriptan, Rizatriptan, Sumatriptan, Zolmitriptan, Aluminium salts, Calcium salts, Ferro salts, Silver salts, Zinc-salte, Amphotericin B, Chlorhexidine, Miconazole, Triamcinolonacetamid, Melatonin, Phenobarbitol, Caffeine, Benzodiazepiner, Hydroxyzine, Meprobamate, Phenothiazine, Buclizine, Brometazine, Cinnarizine, Cyclizine, Difenhydramine, Dimenhydrinate, Buflomedil,
- 15 Amphetamine, Caffeine, Ephedrine, Orlistat, Phenylephedrine, Phenylpropanolamin, Pseudoephedrine, Sibutramin, Ketoconazole, Nitroglycerin, Nystatin, Progesterone, Testosterone, Vitamin B12, Vitamin C, Vitamin A, Vitamin D, Vitamin E, Pilocarpin, Aluminiumaminoacetat, Cimetidine, Esomeprazole, Famotidine, Lansoprazole, Magnesiumoxide, Nizatide and/or Ratinidine or derivatives and
- 20 mixtures thereof.

In an embodiment of the invention, the chewing gum is substantially free of non-biodegradable polymers

- 25 In an embodiment of the invention the at least two or more cyclic esters are selected from the groups of glycolides, lactides, lactones, cyclic carbonates or mixtures thereof.

- In an embodiment of the invention the lactone monomers are chosen from the group
- 30 of  $\epsilon$ -caprolactone,  $\delta$ -valerolactone,  $\gamma$ -butyrolactone, and  $\beta$ -propiolactone. It also includes  $\epsilon$ -caprolactones,  $\delta$ -valerolactones,  $\gamma$ -butyrolactones, or  $\beta$ -propiolactones that have been substituted with one or more alkyl or aryl substituents at any non-carbonyl



carbon atoms along the ring, including compounds in which two substituents are contained on the same carbon atom.

In an embodiment of the invention the carbonate monomer is selected from the group  
5 of trimethylene carbonate, 5-alkyl-1,3-dioxan-2-one, 5,5-dialkyl-1,3-dioxan-2-one,  
or 5-alkyl-5-alkyloxycarbonyl-1,3-dioxan-2-one, ethylene carbonate, 3-ethyl-3-  
hydroxymethyl, propylene carbonate, trimethylolpropane monocarbonate, 4,  
6dimethyl-1, 3-propylene carbonate, 2, 2-dimethyl trimethylene carbonate, and 1, 3-  
dioxepan-2-one and mixtures thereof.

10

In an embodiment of the invention the cyclic ester polymers and their copolymers  
resulting from the polymerization of cyclic ester monomers include, but are not  
limited to : poly (L-lactide) ; poly (D-lactide) ; poly (D, L-lactide) ; poly  
(mesolactide) ; poly (glycolide) ; poly (trimethylenecarbonate) ; poly (epsilon-  
15 caprolactone) ; poly (L-lactide-co-D, L-lactide) ; poly (L-lactide-co-meso-lactide) ;  
poly (L-lactide-co-glycolide) ; poly (L-lactide-co-trimethylenecarbonate) ; poly (L-  
lactide-co-epsilon-caprolactone) ; poly (D, L-lactide-co-meso-lactide) ; poly (D, L-  
lactide-co-glycolide) ; poly (D, L-lactide-co-trimethylenecarbonate) ; poly (D, L-  
lactide-co-epsilon-caprolactone) ; poly (meso-lactide-co-glycolide) ; poly (meso-  
20 lactide-co-trimethylenecarbonate) ; poly (meso-lactide-co-epsilon-caprolactone) ;  
poly (glycolide-cotrimethylenecarbonate) ; poly (glycolide-co-epsilon-caprolactone).

In an embodiment of the invention the chewing gum comprises filler.

25

A chewing gum base formulation may, if desired, include one or more  
fillers/texturisers including as examples, magnesium and calcium carbonate, sodium  
sulphate, ground limestone, silicate compounds such as magnesium and aluminium  
silicate, kaolin and clay, aluminium oxide, silicon oxide, talc, titanium oxide,  
30 mono-, di- and tri-calcium phosphates, cellulose polymers, such as wood, and  
combinations thereof.

In an embodiment of the invention the chewing gum comprises filler in an amount of about 0 to about 50% by weight of the chewing gum, more typically about 10 to about 40% by weight of the chewing gum.

- 5 In an embodiment of the invention the chewing gum comprises at least one coloring agent.

- According to an embodiment of the invention, the chewing gum may comprise color agents and whiteners such as FD&C-type dyes and lakes, fruit and vegetable  
10 extracts, titanium dioxide and combinations thereof. Further useful chewing gum base components include antioxidants, e.g. butylated hydroxytoluene (BHT), butyl hydroxyanisol (BHA), propylgallate and tocopherols, and preservatives.

- 15 In an embodiment of the invention the chewing gum is coated with an outer coating.

In an embodiment of the invention the outer coating is a hard coating.

- In an embodiment of the invention the hard coating is a coating selected from the  
20 group consisting of a sugar coating and a sugarless coating and a combination thereof.

- In an embodiment of the invention the hard coating comprises 50 to 100% by weight of a polyol selected from the group consisting of sorbitol, maltitol, mannitol, xylitol,  
25 erythritol, lactitol and isomalt.

- In an embodiment of the invention the outer coating is an edible film comprising at least one component selected from the group consisting of an edible film-forming agent and a wax.

In an embodiment of the invention the film-forming agent is selected from the group consisting of a cellulose derivative, a modified starch, a dextrin, gelatine, shellac, gum arabic, zein, a vegetable gum, a synthetic polymer and any combination thereof.

- 5 In an embodiment of the invention the outer coating comprises at least one additive component selected from the group consisting of a binding agent, a moisture absorbing component, a film forming agent, a dispersing agent, an antisticking component, a bulking agent, a flavouring agent, a colouring agent, a pharmaceutically or cosmetically active component, a lipid component, a wax  
10 component, a sugar, an acid and an agent capable of accelerating the after-chewing degradation of the degradable polymer.

In an embodiment of the invention the outer coating is a soft coating.

- 15 In an embodiment of the invention the soft coating comprises a sugar free coating agent.

In an embodiment of the invention the chewing gum comprises conventional chewing gum polymers or resins.

20

In an embodiment of the invention the at least one biodegradable polymer comprises at least 5% of the chewing gum polymers.

- 25 In an embodiment of the invention all the biodegradable polymers comprised in the chewing gum comprises at least 25%, preferably at least 50% of the chewing gum polymers.

- 30 In an embodiment of the invention the biodegradable polymers comprised in the chewing gum comprises at least 80%, preferably at least 90% of the chewing gum polymers.

In an embodiment of the invention the chewing gum comprises

said at least one biodegradable polyester copolymer forming a plasticizer of the chewing gum and at least one non-biodegradable conventional elastomer.

According to the invention, a biodegradable polymer according to the invention may  
5 form a substitute of a conventional natural or synthetic resin.

In an embodiment of the invention the chewing gum comprises  
the at least one biodegradable polyester copolymer forming an elastomer of the  
chewing gum and at least one non-biodegradable conventional natural or synthetic  
10 resin.

According to the invention, a biodegradable polymer according to the invention may  
form a substitute of a conventional low or high molecular weight elastomer.

15 In an embodiment of the invention said chewing gum comprises

at least one biodegradable elastomer in the amount of about 0.5 to about 70% wt of  
the chewing gum,

20 at least one biodegradable plasticizer in the amount of about 0.5 to about 70% wt of  
the chewing gum and

at least one chewing gum ingredient chosen from the groups of softeners, sweeteners,  
flavoring agents, active ingredients and fillers in the amount of about 2 to about 80%  
25 wt of the chewing gum.

In an embodiment of the invention edible polyesters may be applied as a degradable  
chewing gum polymer.

30 Edible polyesters are obtained by esterification of at least one alcohol and one acid.

The edible polyester is produced by condensation polymerization reaction of at least one alcohol chosen from the group of trihydroxyl alcohol and dihydroxyl alcohol, and at least one acid chosen from the group consisting of dicarboxylic acid and tricarboxylic acid.

5

It is possible to use edible or food grade materials. Because the starting acids and alcohols are food grade materials the resultant polymers is edible.

Alcohols: Glycerol, propylene glycol, 1,3 butylene diol

10

Acids: Citric acid, fumaric acid, adipic acid, malic acid, succinic acid, suberic acid, sebacic acid, dodecanedioic acid, glucaric acid, glutamic acid, glutaric, azelaic acid, tartaric acid

15 Edible polyesters can replace both elastomers and elastomer plasticizers and form 1-80% of the gum base.

### Drawings

The invention will now be described with reference to the drawings of which

20

fig. 1 illustrates  $G'$  (storage modulus) versus oscillation torque for chewing gums 1002, 1003 and 1004, all containing 3% lecithin and where

fig. 2 illustrates  $\tan(\delta)$  versus oscillation torque for chewing gums 1002, 25 1003 and 1004, all containing 3% lecithin,

### Detailed description

30 In the present context the terms environmentally or biologically degradable polymer compounds refers to chewing gum base components which, after dumping the chewing gum, is capable of undergoing a physical, chemical and/or biological

degradation whereby the dumped chewing gum waste becomes more readily removable from the site of dumping or is eventually disintegrated to lumps or particles which are no longer recognisable as being chewing gum remnants. The degradation or disintegration of such degradable polymers can be effected or induced  
5 by physical factors such as temperature, light, moisture, by chemical factors such as hydrolysis caused by a change in pH or by the action of enzymes capable of degrading the polymers. In other useful embodiments all of the polymer components of the gum base are environmentally degradable or biodegradable polymers.

10 Preferably, the ultimate degradation products are carbon dioxide, methane and water.

According to a preferred definition of biodegradability according to the invention biodegradability is a property of certain organic molecules whereby, when exposed to the natural environment or placed within a living organism, they react through an  
15 enzymatic or microbial process, often in combination with a pure chemical process such as hydrolysis, to form simpler compounds, and ultimately, carbon dioxide, nitrogen oxides, and water.

Accordingly, suitable examples of additional environmentally or biologically degradable chewing gum base polymers which can be applied in accordance with the gum  
20 base of the present invention include degradable polyesters, polycarbonates, polyester amides, polypeptides, homopolymers of amino acids such as polylysine, and proteins including derivatives hereof such as e.g. protein hydrolysates including a zein hydrolysate. Particularly useful compounds of this type include polyester  
25 polymers obtained by the polymerisation of one or more cyclic esters such as lactide, glycolide, trimethylene carbonate,  $\delta$ -valerolactone,  $\beta$ -propiolactone and  $\epsilon$ -caprolactone. Such degradable polymers may be homopolymers or copolymers, including block-polymers.

30 Unless otherwise indicated, as used herein, the term "molecular weight" means number average molecular weight ( $M_n$ ).

In the following examples, two different conventional chewing gumbases are made. The main difference between the two gumbases 101 and 102 of EXAMPLE 6 is the difference in molecular weight of the high molecular weight elastomer, i.e. the butylrubber.

5

The following examples are provided for the purpose of demonstrating the different properties of conventional and non-biodegradable elastomers when incorporated in a gumbase and a chewing gum formulation. The variable parameters, i.e. the molecular weight of the applied high molecular weight elastomers, and chewing gum ingredients has been kept low in order to highlight the actual difference between the chewing gums made on a conventional basis and chewing gum made on the basis of biodegradable polymers.

10

In EXAMPLE 6, gumbases 103 and 104 have been made by means of the same process, but the conventional high molecular weight elastomer has now been exchanged with a biodegradable substitute having almost the same molecular weight.

15

In the EXAMPLE 6, gumbase 105 has the molecular weight of the biodegradable high molecular weight elastomer has been further increased.

20

#### EXAMPLE 1

##### Preparation of resin

25

A resin sample was produced using a cylindrical glass, jacketed 10 L pilot reactor equipped with glass stir shaft and Teflon stir blades and bottom outlet. Heating of the reactor contents was accomplished by circulation of silicone oil, thermostated to 130°C, through the outer jacket. D,L-lactide (4.877 kg, 33.84 mol) was charged to the reactor and melted by heating to 140°C for 6 h. After the D,L-lactide was completely molten, the temperature was reduced to 130°C, and stannous octoate (1.79 g,  $4.42 \times 10^{-3}$  mol), 1,2-propylene glycol (79.87 g, 1.050 mol), and  $\epsilon$ -

30

caprolactone (290.76 g, 2.547 mol) were charged to the reactor. After the mixture became homogeneous, stirring was continued for 24 h at 130°C. At the end of this time, the bottom outlet was opened, and molten polymer was allowed to drain into a Teflon-lined paint can.

- 5 Characterization of the product indicated  $M_n = 5,700$  g/mol and  $M_w = 7,100$  g/mol (gel permeation chromatography with online MALLS detector) and  $T_g = 30.7^\circ\text{C}$  (DSC, heating rate  $10^\circ\text{C}/\text{min}$ ).

## EXAMPLE 2

10

### Preparation of LMWE elastomer

- A 515 g LMWE sample was synthesized within a dry  $\text{N}_2$  glove box, as follows. Into a 500 mL resin kettle equipped with overhead mechanical stirrer, 0.73 g 1,2-propane diol (3.3mL of a 22.0%(w/v) solution in methylene chloride), and 0.152 g  $\text{Sn}(\text{Oct})_2$  (3.56 ml of a 4.27% (w/v) solution in methylene chloride) were charged under dry  $\text{N}_2$  gas purge. The methylene chloride was allowed to evaporate under the  $\text{N}_2$  purge for 15 min. Then  $\epsilon$ -caprolactone (300g, 2.63 mol) and  $\delta$ -valerolactone (215 gm, 2.15 mol) were added. The resin kettle was submerged in a  $130^\circ\text{C}$  constant temperature oil bath and stirred for 14 h. Subsequently the kettle was removed from the oil bath and allowed to cool at room temperature. The solid, elastic product was removed in small pieces using a knife, and placed into a plastic container.

- 20 Characterization of the product indicated  $M_n = 59,900$  g/mol and  $M_w = 74,200$  g/mol (gel permeation chromatography with online MALLS detector) and  $T_g = -70^\circ\text{C}$  (DSC, heating rate  $10^\circ\text{C}/\text{min}$ ).

## EXAMPLE 3

- 30 Preparation of HMWE



A HMWE sample according to the invention was synthesized in a dry N<sub>2</sub> glove box, as follows. Into a 500 mL resin kettle equipped with overhead mechanical stirrer was charged 0.037 g Sn(Oct)<sub>2</sub> (3.4 mL of a 1.10% (w/v) solution in methylene chloride) under dry N<sub>2</sub> gas purge. The methylene chloride was allowed to evaporate under the N<sub>2</sub> purge for 15 min. Then, pentaerythritol (0.210 g, 1.54 x 10<sup>-3</sup> mol), ε-caprolactone (79.0g, 0.692 mol), TMC(8.0 g, 0.078 mol) and δ-valerolactone (38.0 g, 0.380 mol) were added. The resin kettle was submerged in a 130°C constant temperature oil bath and stirred for 14 h. Subsequently the kettle was removed from the oil bath and allowed to cool at room temperature. The solid, elastic product was removed in small pieces using a knife, and placed into a plastic container.

Characterization of the product indicated  $M_n = 64,600$  g/mol and  $M_w = 165,200$  g/mol (gel permeation chromatography with online MALLS detector) and  $T_g = -66^\circ\text{C}$  (DSC, heating rate 10°C/min).

15

#### EXAMPLE 4

##### Preparation of high HMWE

A HMWE sample was synthesized in a dry N<sub>2</sub> glove box, as follows. Into a 500 mL resin kettle equipped with overhead mechanical stirrer was charged 0.037 g Sn(Oct)<sub>2</sub> (2.4 mL of a 1.54% (w/v) solution in methylene chloride) under dry N<sub>2</sub> gas purge. The methylene chloride was allowed to evaporate under the N<sub>2</sub> purge for 15 min. Then, pentaerythritol (0.068 g, 4.99 x 10<sup>-4</sup> mol), ε-caprolactone (68.0g, 0.596 mol), TMC (7.0 g, 0.069 mol), and δ-valerolactone (33.0 g, 0.33 mol) were added. The resin kettle was then submerged in a 130°C constant-temperature oil bath and stirred for about 2 - 2.5 h, at which time the mass solidified and could no longer be stirred. The reacting mass was then maintained at 130°C for an additional 11.5 - 12 h for a total reaction time of 14 h. Subsequently the kettle was removed from the oil bath and allowed to cool to room temperature. The solid, elastic product was removed in small pieces using a knife, and placed into a plastic container.

Characterization of the product indicated  $M_n = 113,900$  g/mol and  $M_w = 369,950$  g/mol (gel permeation chromatography with online MALLS detector).

## 5 EXAMPLE 5

### Preparation of a high HMWE

A 219 g HMWE sample was synthesized as follows in a dry N<sub>2</sub> glove box. Into a 500  
10 mL resin kettle, equipped with overhead mechanical stirrer was charged Stannous  
ethoxide (0.077 g,  $3.69 \times 10^{-4}$  mol),  $\epsilon$ -caprolactone (137 g, 1.2 mol), and TMC (82  
g, 0.804 mol) were added. The resin kettle was then submerged in a 130°C constant-  
temperature oil bath and stirred for 14 h. Subsequently the kettle was removed from  
the oil bath and allowed to cool to room temperature. The solid, elastic product was  
15 removed in small pieces using a knife, and placed into a plastic container.  
Characterization of the product indicated  $M_n = 254,900$  g/mol (gel permeation  
chromatography with online MALLS detector).

## 20 EXAMPLE 6

### Preparation of gumbases

All the gumbases are prepared with following basic formulation:

25

<u>Ingredients</u>	<u>Percent by weight</u>
Elastomer HMWE	20
Elastomer LMWE	40
30 Resin	40

No	Type	Elastomer HMWE	Elastomer LMWE	Resin
101	Standard	Polyisobutylene Mn =73,000	Polyisobutylene Mn =30.000	Polyvinylacetate Mn =5000
102	Standard	Butylrubber Mn = 117,000	Polyisobutylene Mn = 30.000	Polyvinylacetate Mn = 5000
103	Biodegradable	Elastomer polymer from example 3	Elastomer polymer from example 2	Resin polymer from example 1
104	Biodegradable	Elastomer polymer from example 4	Elastomer polymer from example 2	Resin polymer from example 1
105	Biodegradable	Elastomer polymer from example 5	Elastomer polymer from example 2	Resin polymer from example 1

Table 1: Gumbase preparation

The gumbases are prepared as follows:

5

HMWE elastomer is added to a mixing kettle provided with mixing means like e.g. horizontally placed Z-shaped arms. The kettle had been preheated for 15 minutes to a temperature of about 60-80°C. The rubber is broken into small pieces and softened with mechanical action on the kettle.

10

The resin is slowly added to the elastomer until the mixture becomes homogeneous. The remaining resin is then added to the kettle and mixed for 10-20 minutes. The LMWE elastomer is added and mixed for 20-40 minutes until the whole mixture becomes homogeneous.

15

The mixture is then discharged into the pan and allowed to cool to room temperature from the discharged temperature of 60-80°C, or the gumbase mixture is used directly for chewing gum by adding all chewing gum components in an appropriate order under continuous mixing.

20

## EXAMPLE 7

**Preparation of Chewing gum**

- 5 The gumbases listed in table 1 were mixed into a final chewing gum during addition of different chewing gum ingredients as illustrated in the table below.

	1001	1002	1003	1004	1005	1006	1007
Parts (weight)	%	%	%	%	%	%	%
Gumbase	40% of 101	40% of 102	40% of 103	40% of 103	40% of 104	40% of 104	40% of 105
Sorbitol powder	45.6	45.6	45.6	47.3	45.6	44.6	45.6
Lycasin	3	3	3	3	3	3	3
Peppermint oil	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Menthol (crystal)	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Aspartame	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Acesulfame	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Xylitol	6	6	6	6	6	6	6
Fat	0	0	0	0.3	0	4	0

Lecithin	3	3	3	1.0	3	0	3
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Table 2: Preparation of chewing gum

The gum bases used in the above chewing gum formulations are relatively hard and the chewing gum system needs additional softening in order to get an acceptable texture.

The two standard formulation 1001 and 1002, containing HMWE elastomers with Mn of 73,000 and 117,000 showed improved texture with 3% lecithin. A biodegradable gum base (103) including a polymer with Mn of 65.000 (EXAMPLE 3) was tested in chewing gum with 3% lecithin (1003), the chewing gum became very soft, and it almost dissolved while chewing. The same gumbase (103) was used in a chewing gum formulation (1004) where the softening system comprises 1% lecithin and 0.3% fat, this formulation was also too soft.

It can therefore be concluded that the biodegradable gumbases appears to be less resistant to softeners and emulsifiers.

Chewing gum formulation 1005 and 1006 was prepared with a new biodegradable gumbase (104) containing a polymer with a higher Mn of 114.000 (EXAMPLE 4). When adding 3% lecithin or 4% fat the chewing gum has a pleasant and acceptable texture compared to conventional chewing gum formulations.

Chewing gum formulation 1007 was made with a biodegradable gumbase 105, where a polymer with Mn of 350.000 was used (EXAMPLE 5). When adding 3% lecithin the texture is harder but acceptable.

Hence, an increase of molecular weight results in an increased polymer resistance to chewing gum ingredients such as for instance softeners and fat.

## EXAMPLE 8

An experiment was set up in order to test different chewing gum formulations containing 3% lecithin.

- 5 1001 and 1002 are two standard formulations containing elastomers with Mn of 73,000 and 117,000.

- 1003 is a 100% biodegradable formulation containing elastomer polymer Mn of 65,000 and 1004 is a 100% biodegradable formulation containing elastomer polymer  
10 with Mn of 114,000.

- The gum centres were chewed in a chewing machine (CF Jansson). The chewing frequency was set to 1 Hz, a pH buffer was used as saliva and the temperature was set at 37°C. The chewing time was set to 30 seconds. After chewing, the chewed cud  
15 was measured on a rheometer, type AR1000 from TA Instruments. The oscillation measurement is performed at a stress within the linear viscoelastic region and a temperature of 37°C with a parallel plate system (d=2.0 cm, hatched). G', and tan delta vs. shear rate.

- 20 The results are summarised in fig.1 and fig. 2, and as it appears, the biodegradable formulations containing 3% lecithin show different rheological behavior. The low Mn of 65,000 (1003) is very soft and less elastic compared to the formulation with high Mn (1004).

This is confirming the sensorial evaluation described in the above EXAMPLE 7.

Patent Claims

1. Chewing gum comprising at least one biodegradable polymer, wherein the molecular weight of said biodegradable polymer is at least 105000 g/mol (Mn).  
5
2. Chewing gum according to claim 1, wherein the molecular weight of said at least one biodegradable polymer is at least 150000 g/mol (Mn).
3. Chewing gum according to claim 1 or 2, wherein the molecular weight of said at  
10 least one biodegradable polymer is within the range of 105000 g/mol (Mn) to 1000000 g/mol (Mn).
4. Chewing gum according to any of the claims 1 to 3, wherein the molecular weight of said at least one biodegradable polymer is within the range of 105000 g/mol (Mn)  
15 to 500000 g/mol (Mn).
5. Chewing gum according to any of the claims 1 to 4, wherein the molecular weight of said at least one biodegradable polymer is within the range of 105000 g/mol (Mn) to 350000 g/mol (Mn).  
20
6. Chewing gum according to any of the claims 1 to 5, wherein the molecular weight of said at least one biodegradable polymer is within the range of 105000 g/mol (Mn) to 250000 g/mol (Mn).
- 25 7. Chewing gum according to any of the claims 1 to 6, wherein the molecular weight of said at least one biodegradable polymer is less than 2000000 g/mol (Mn).
8. Chewing gum according to any of the claims 1 to 7, wherein the polydispersity of said at least one biodegradable polymer is within the range of 1 to 5.
- 30 9. Chewing gum according to any of the claims 1 to 8, wherein the polydispersity of said at least one biodegradable polymer is within the range of 1 to 2.5.

10. Chewing gum according to any of the claims 1 to 9, wherein the at least one biodegradable polymer comprises at least 25% of the chewing gum polymers, preferably at least 50% of the chewing gum polymers.
- 5 11. Chewing gum according to any of the claims 1 to 10, wherein all the biodegradable polymers comprised in the chewing gum comprises at least 25%, preferably at least 90% of the chewing gum polymers.
- 10 12. Chewing gum according to any of the claims 1 to 10, wherein all the biodegradable polymers comprised in the chewing gum comprises at least 80%, preferably at least 95% of the chewing gum polymers.
- 15 13. Chewing gum according to any of the claims 1 to 12, wherein the chewing gum is substantially free of non-biodegradable polymers.
14. Chewing gum according to any of claims 1-13, wherein said chewing gum ingredients comprises flavoring agents.
- 20 15. Chewing gum according to any of claims 1-14, wherein said flavoring agents comprises natural and synthetic flavorings in the form of natural vegetable components, essential oils, essences, extracts, powders, including acids and other substances capable of affecting the taste profile.
- 25 16. Chewing gum according to any of claims 1-15, wherein said chewing gum comprises flavor in an amount of 0.01 to about 30 wt%, said percentage being based on the total weight of the chewing gum
- 30 17. Chewing gum according to any of claims 1-16, wherein said chewing gum comprises flavor in an amount of 0.2 to about 4 wt%, said percentage being based on the total weight of the chewing gum



18. Chewing gum according to any of claims 1-17, wherein said flavor comprises water soluble ingredients.
19. Chewing gum according to any of claims 1-18, wherein  
5 said water soluble flavor comprises acids.
20. Chewing gum according to any of claims 1-19, wherein said flavor comprises water insoluble ingredients.
- 10 21. Chewing gum according to any of claims 1-20, wherein said chewing gum ingredients comprises sweeteners.
22. Chewing gum according to any of claims 1-21, wherein said sweetener comprises bulk sweeteners  
15
23. Chewing gum according to any of claims 1-22, wherein the chewing gum comprises bulk sweeteners in the amount of about 5 to about 95% by weight of the chewing gum, more typically about 20 to about 80% by weight of the chewing gum.  
20
24. Chewing gum according to any of claims 1-23, wherein said sweetener comprises high intensity sweeteners
25. Chewing gum according to any of claims 1-24, wherein the high intensity  
25 sweeteners comprise sucralose, aspartame, salts of acesulfame, alitame, saccharin and its salts, cyclamic acid and its salts, glycyrrhizin, dihydrochalcones, thaumatin, monellin, sterioside, alone or in combination
26. Chewing gum according to any of claims 1-25,  
30 wherein the chewing gum comprises high intensity sweeteners in an amount of about 0 to about 1% by weight of the chewing gum, more typically about 0.05 to about 0.5% by weight of the chewing gum.

27. Chewing gum according to any of claims 1-26,  
wherein the chewing gum comprises at least one softener.
- 5 28. Chewing gum according to any of claims 1-27,  
wherein the at least one softener comprises tallow, hydrogenated tallow,  
hydrogenated and partially hydrogenated vegetable oils, cocoa butter, glycerol  
monostearate, glycerol triacetate, lecithin, mono-, di- and triglycerides, acetylated  
monoglycerides, fatty acids - such as stearic, palmitic, oleic and linoleic acids  
10 mixtures thereof.
29. Chewing gum according to any of claims 1-28,  
wherein the chewing gum comprises softeners in the amount of about 0 to about 18%  
by weight of the chewing gum, more typically about 0 to about 12% by weight of the  
15 chewing gum.
30. Chewing gum according to any of claims 1-29, wherein said chewing gum  
ingredients comprise active ingredients.
- 20 31. Chewing gum according to any of claims 1-30, wherein  
said active ingredients being selected from the group of: Acetaminophen,  
Acetylsalicylsyre Buprenorphine Bromhexin Celcoxib Codeine, Diphenhydramin,  
Diclofenac, Etoricoxib, Ibuprofen, Indometacin, Ketoprofen, Lumiracoxib,  
Morphine, Naproxen, Oxycodon, Parecoxib, Piroxicam, Pseudoefedrin, Rofecoxib,  
25 Tenoxicam, Tramadol, Valdecocix, Calciumcarbonat, Magaldrate, Disulfiram,  
Bupropion, Nicotine, Azithromycin, Clarithromycin, Clotrimazole, Erythromycin,  
Tetracycline, Granisetron, Ondansetron, Prometazin, Tropisetron, Brompheniramine,  
Ceterizin, Ieco-Ceterizin, Chlorcyclizine, Chlorpheniramin, Chlorpheniramin,  
Difenhydramine, Doxylamine, Fenofenadin, Guaifenesin, Loratidin, des-Loratidin,  
30 Phenyltoloxamine, Promethazin, Pyridamine, Terfenadin, Troxerutin, Methyldopa,  
Methylphenidate, Benzalcon. Chloride, Benzeth. Chloride, Cetylpyrid. Chloride,  
Chlorhexidine, Ecabet-sodium, Haloperidol, Allopurinol, Colchicine, Theophylline,

- Propanolol, Prednisolone, Prednisone, Fluoride, Urea, Miconazole, Actot,  
Glibenclamide, Glipizide, Metformin, Miglitol, Repaglinide, Rosiglitazone,  
Apomorphin, Cialis, Sildenafil, Vardenafil, Diphenoxylate, Simethicone, Cimetidine,  
Famotidine, Ranitidine, Ratinidine, cetrizin, Loratadine, Aspirin, Benzocaine,  
5 Dextrometorphan, Ephedrine, Phenylpropanolamine, Pseudoephedrine, Cisapride,  
Domperidone, Metoclopramide, Acyclovir, Dioctylsulfosucc., Phenolphthalein,  
Almotriptan, Eletriptan, Ergotamine, Migea, Naratriptan, Rizatriptan, Sumatriptan,  
Zolmitriptan, Aluminium salts, Calcium salts, Ferro salts, Silver salts, Zinc-salte,  
Amphotericin B, Chlorhexidine, Miconazole, Triamcinolonacetoneid, Melatonine,  
10 Phenobarbitol, Caffeine, Benzodiazepiner, Hydroxyzine, Meprobamate,  
Phenothiazine, Buclizine, Brometazine, Cinnarizine, Cyclizine, Difenhydramine,  
Dimenhydrinate, Buflomedil, Amphetamine, Caffeine, Ephedrine, Orlistat,  
Phenylephedrine, Phenylpropanolamin, Pseudoephedrine, Sibutramin, Ketoconazole,  
Nitroglycerin, Nystatin, Progesterone, Testosterone, Vitamin B12, Vitamin C,  
15 Vitamin A, Vitamin D, Vitamin E, Pilocarpin, Aluminiumaminoacetat, Cimetidine,  
Esomeprazole, Famotidine, Lansoprazole, Magnesiumoxide, Nizatide and/or  
Ratinidine or derivates and mixtures thereof.

32. Chewing gum according to any of claims 1-31, wherein the chewing gum is  
20 substantially free of non-biodegradable polymers

33. Chewing gum according to any of claims 1-32, wherein the at least one  
biodegradable polyester copolymer obtained by the polymerization of one or more  
cyclic esters by ring-opening and where at least one of the cyclic esters are selected  
25 from the groups of glycolides, lactides, lactones, cyclic carbonates or mixtures  
thereof.

34. Chewing gum according to any of claims 1-33, wherein lactone monomers are  
chosen from the group of  $\epsilon$ -caprolactone,  $\delta$ -valerolactone,  $\gamma$ -butyrolactone, and  $\beta$ -  
30 propiolactone. It also includes  $\epsilon$ -caprolactones,  $\delta$ -valerolactones,  $\gamma$ -butyrolactones, or  
 $\beta$ -propiolactones that have been substituted with one or more alkyl or aryl

substituents at any non-carbonyl carbon atoms along the ring, including compounds in which two substituents are contained on the same carbon atom.

35. Chewing gum according to any of claims 1-34,  
5 wherein the carbonate monomer is selected from the group of trimethylene carbonate, 5-alkyl-1,3-dioxan-2-one, 5,5-dialkyl-1,3-dioxan-2-one, or 5-alkyl-5-alkyloxycarbonyl-1,3-dioxan-2-one, ethylene carbonate, 3-ethyl-3-hydroxymethyl, propylene carbonate, trimethylolpropane monocarbonate, 4,6-dimethyl-1,3-propylene carbonate, 2,2-dimethyl trimethylene carbonate, and 1,3-dioxepan-2-one  
10 and mixtures thereof.

36. Chewing gum according to any of claims 1-35,  
wherein cyclic ester polymers and their copolymers resulting from the polymerization of cyclic ester monomers include, but are not limited to: poly (L-  
15 lactide) ; poly (D-lactide) ; poly (D, L-lactide) ; poly (mesolactide) ; poly (glycolide) ; poly (trimethylenecarbonate) ; poly (epsilon-caprolactone) ; poly (L-lactide-co-D, L-lactide) ; poly (L-lactide-co-meso-lactide) ; poly (L-lactide-co-glycolide) ; poly (L-lactide-co-trimethylenecarbonate) ; poly (L-lactide-co-epsilon-caprolactone) ; poly (D, L-lactide-co-meso-lactide) ; poly (D, L-  
20 lactide-co-glycolide) ; poly (D, L-lactide-co-trimethylenecarbonate) ; poly (D, L-lactide-co-epsilon-caprolactone) ; poly (meso-lactide-co-glycolide) ; poly (meso-lactide-co-trimethylenecarbonate) ; poly (meso-lactide-co-epsilon-caprolactone) ; poly (glycolide-cotrimethylenecarbonate) ; poly (glycolide-co-epsilon-caprolactone).

- 25  
37. Chewing gum according to any of claims 1-36,  
wherein the chewing comprises filler.

38. Chewing gum according to any of claims 1-37,  
30 wherein the chewing gum comprises filler in an amount of about 0 to about 50% by weight of the chewing gum, more typically about 10 to about 40% by weight of the chewing gum.

39. Chewing gum according to any of claims 1-38,  
wherein the chewing gum comprises at least one coloring agent.
- 5 40. Chewing gum according to any of claims 1-39, where the chewing gum is coated  
with an outer coating.
41. Chewing gum according to any of claims 1-40, wherein the outer coating is a  
hard coating.
- 10 42. Chewing gum according to any of claims 1-41, wherein the hard coating is a  
coating selected from the group consisting of a sugar coating and a sugarless coating  
and a combination thereof.
- 15 43. Chewing gum according to any of claims 1-42, wherein the hard coating com-  
prises 50 to 100% by weight of a polyol selected from the group consisting of  
sorbitol, maltitol, mannitol, xylitol, erythritol, lactitol and isomalt.
44. Chewing gum according to any of claims 1-43, wherein the outer coating is an  
20 edible film comprising at least one component selected from the group consisting of  
an edible film-forming agent and a wax.
45. Chewing gum according to any of claims 1-44, wherein the film-forming agent is  
selected from the group consisting of a cellulose derivative, a modified starch, a  
25 dextrin, gelatine, shellac, gum arabic, zein, a vegetable gum, a synthetic polymer and  
any combination thereof.
46. Chewing gum according to any of claims 1-45, wherein the outer coating  
comprises at least one additive component selected from the group consisting of a  
30 binding agent, a moisture absorbing component, a film forming agent, a dispersing  
agent, an antisticking component, a bulking agent, a flavouring agent, a colouring  
agent, a pharmaceutically or cosmetically active component, a lipid component, a

wax component, a sugar, an acid and an agent capable of accelerating the after-chewing degradation of the degradable polymer.

47. Chewing gum according to any of claims 1-46, wherein the outer coating is a soft  
5 coating.

48. Chewing gum according to any of claims 1-47, wherein the soft coating comprises a sugar free coating agent.

10 49. Chewing gum according to any of the claims 1-48, wherein said chewing gum comprises

at least one biodegradable elastomer in the amount of about 0.5 to about 70% wt of the chewing gum,

15

at least one biodegradable plasticizer in the amount of about 0.5 to about 70% wt of the chewing gum and

20 at least one chewing gum ingredient chosen from the groups of softeners, sweeteners, flavoring agents, active ingredients and fillers in the amount of about 2 to about 80% wt of the chewing gum.

50. Method of increasing the robustness of chewing gum comprising at least one biodegradable polymer by increasing the molecular weight of the at least one  
25 biodegradable polymer.

51. Method of increasing the robustness according to claim 50, whereby the molecular weight of said biodegradable polymer is adjusted to be at least 105000 g/mol (Mn).

30

52. Method of increasing the robustness according to claim 50 or 51, whereby the molecular weight of said biodegradable polymer is adjusted to be at least 150000 g/mol (Mn).
- 5 53. Method of increasing the robustness according to any of the claims 50 -52, whereby the molecular weight of said biodegradable polymer is adjusted to be at least 250000 g/mol (Mn).
54. Method of increasing the robustness according to any of the claims 50 -53,  
10 whereby the molecular weight of said at least one biodegradable polymer is adjusted to be within the range of 105000 g/mol (Mn) to 500000 g/mol (Mn).
55. Method of increasing the robustness according to any of the claims 50 -52, whereby the molecular weight of said at least one biodegradable polymer is adjusted  
15 to within the range of 105000 g/mol (Mn) to 350000 g/mol (Mn).
56. Method of increasing the robustness according to any of the claims 50 -55, whereby the molecular weight of said at least one biodegradable polymer is adjusted within the range of 105000 g/mol (Mn) to 250000 g/mol (Mn).  
20
57. Method of increasing the robustness according to any of the claims 50 -56, whereby the molecular weight of said at least one biodegradable polymer is adjusted to be less than 2000000 g/mol (Mn).
- 25 58. Gum base according to any of claims 1-57.

1/2

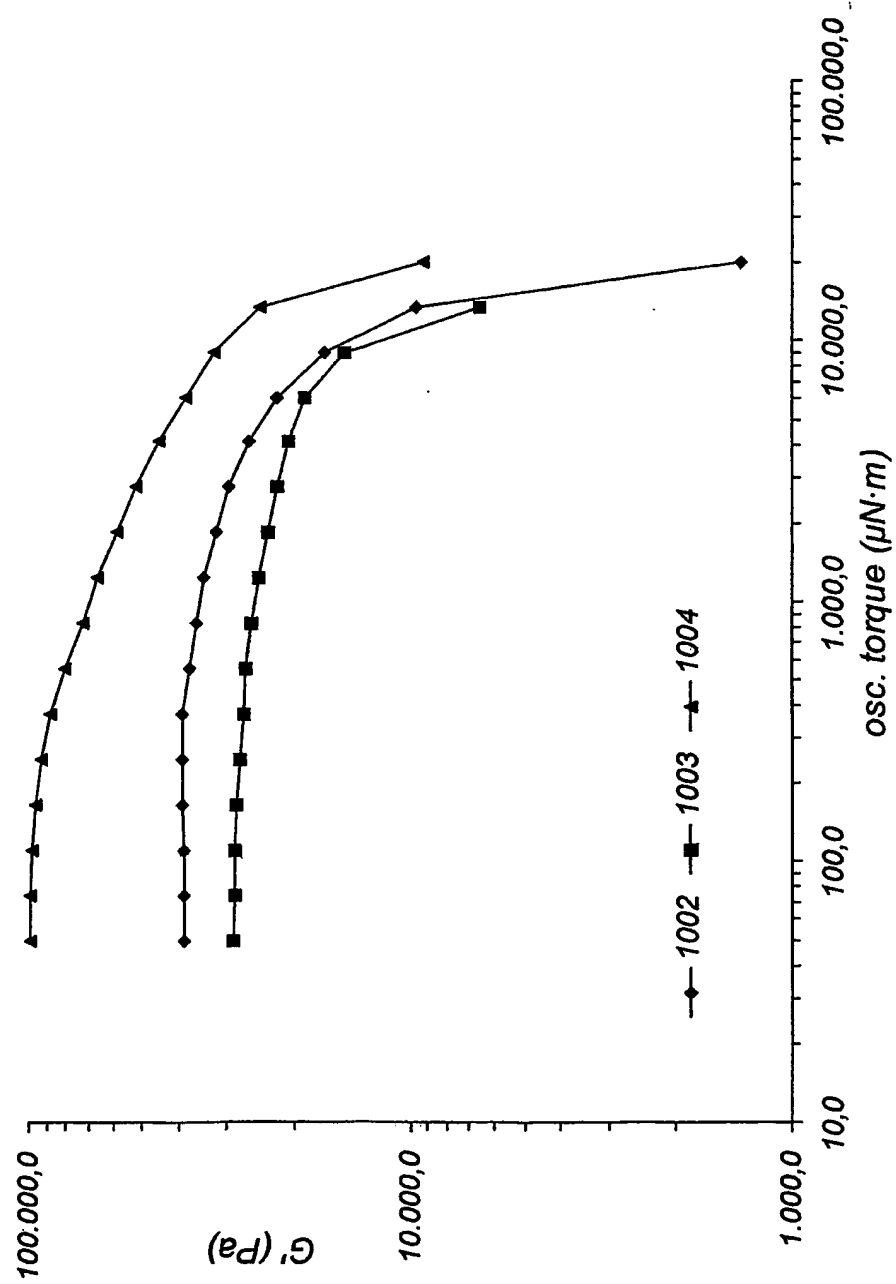


Fig. 1



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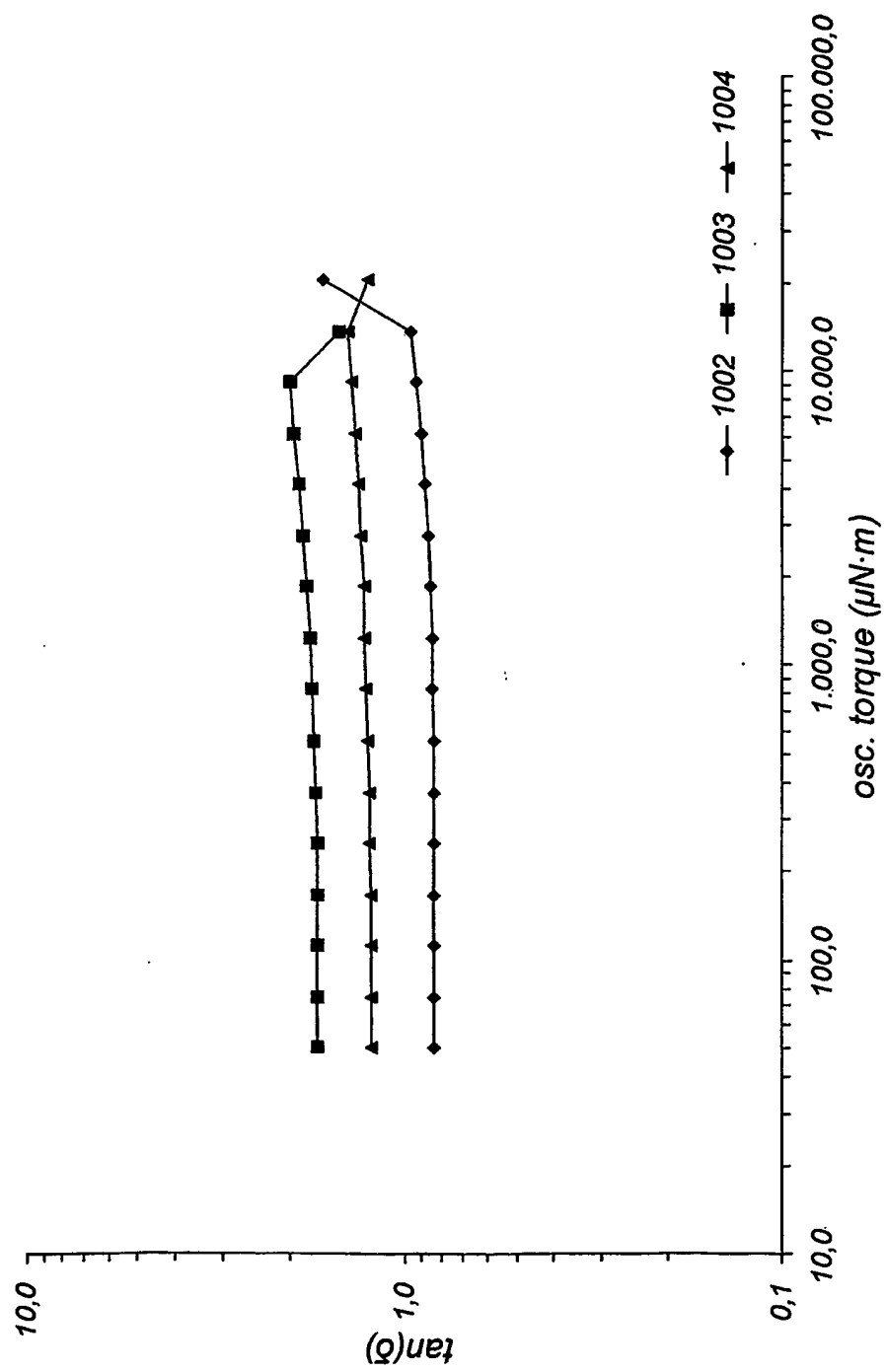


Fig. 2

# INTERNATIONAL SEARCH REPORT

International Application No  
PCT/DK/00625

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 A23G3/30

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 A23G

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 711 506 A (UNIV GRONINGEN) 15 May 1996 (1996-05-15) the whole document ---	1-58
X	WO 01 47368 A (PATEL BHARAT KANAIYALAL ;GOLDBERG DANIEL (US); EATON ROBERT FRANCI) 5 July 2001 (2001-07-05) abstract page 10, line 22 -page 13, line 8 page 19, line 15 - line 21 ---	1-58
A	US 3 800 006 A (KATAYAMA S ET AL) 26 March 1974 (1974-03-26) the whole document ---	1-58
A	WO 00 19837 A (LI WEISHENG ;WRIGLEY W M JUN CO (US)) 13 April 2000 (2000-04-13) the whole document -----	1-58

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

### \* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

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"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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Date of the actual completion of the international search

9 May 2003

Date of mailing of the international search report

18. 06. 2003

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# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/DK92/00625

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